Science of the Total Environment 625 (2018) 872–884

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

# Internal phosphorus loading from sediments causes seasonal nitrogen limitation for harmful algal blooms

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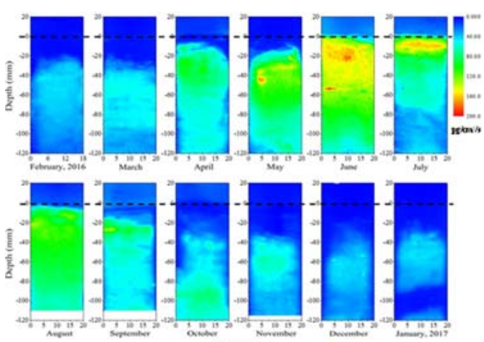
## HIGHLIGHTS

- High-resolution sampling showed seasonal variations in distribution of mobile P in sediments.
- SRP diffusion flux at the sediment-water interface varied from −0.01 to 6.76 mg/m$^2$/d.
- Spatial-temporal variation in mobile P was controlled by microbe-mediated Fe redox cycling.
- Internal P loading accounted for 54% of increased water column TP during the prebloom-bloom period.
- Internal P loading played a major role in causing seasonal nitrogen limitation for HABs.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

*Article history:*
Received 4 November 2017
Received in revised form 31 December 2017
Accepted 31 December 2017
Available online xxxx

Editor: F.M. Tack

*Keywords:*
Lake sediment
Mobile phosphorus
Eutrophication
High-resolution sampling
Iron redox cycle
Harmful algal blooms

## ABSTRACT

It is proposed that the internal loading of phosphorus (P) from sediments plays an important role in seasonal nitrogen (N) limitation for harmful algal blooms (HABs), although there is a lack of experimental evidence. In this study, an eutrophic bay from the large and shallow Lake Taihu was studied for investigating the contribution of internal P to N limitation over one-year field sampling (February 2016 to January 2017). A prebloom-bloom period was identified from February to August according to the increase in Chl*a* concentration in the water column, during which the ratio of total N to total P (TN/TP) exponentially decreased with month from 43.4 to 7.4. High-resolution dialysis (HR-Peeper) and diffusive gradients in thin films (DGT) analysis showed large variations in the vertical distribution of mobile P (SRP and DGT-labile P) in sediments, resulting in the SRP diffusion flux at the sediment-water interface ranging from −0.01 to 6.76 mg/m$^2$/d (minus sign denotes downward flux). Significant and linear correlations existed between SRP and soluble Fe(II) concentrations in pore water, reflecting that the spatial-temporal variation in mobile P was controlled by microbe-mediated Fe redox cycling. Mass estimation showed that the cumulative flux of SRP from sediments accounted for 54% of the increase in TP observed in the water column during the prebloom-bloom period. These findings are supported by the significantly negative correlation ($p < 0.01$) observed between sediment SRP flux and water column TN/TP during the same period. Overall, these results provide solid evidence for the major role of internal P loading in causing N limitation during the prebloom-bloom period.

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https://doi.org/10.1016/j.scitotenv.2017.12.348
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## 1. Introduction

Harmful algal blooms (HABs) are occurring worldwide with increasing magnitude and frequency. They seriously threaten the balance of lake ecosystems, posing a risk to public health by affecting drinking water supplies, food security, and recreational use (Brooks et al. 2016). HABs are an established indicator of water eutrophication, which is caused by the over-enrichment of anthropogenic nutrients in water bodies (Paerl and Otten 2013). Nitrogen (N) and phosphorus (P) are key nutrients in eutrophication; they play a dominant role in the promotion of HABs with significantly increased concentrations in freshwater ecosystems (Peñuelas et al. 2013; Schindler et al. 2016; Tong et al. 2017).

During previous decades, P was regarded as the prime limiting nutrient in freshwater ecosystems (Schelske 2009; Schindler et al. 2008, 2016), based on the evidence from multiscale experiments including long-term case studies and multi-year whole lake assessments (Ho and Michalak 2017; Schindler et al. 2008, 2016). Accordingly, P limitation has received more research attention than N limitation, encouraging the development of effective control measures for P input, while the control of N is assumed to be offset by cyanobacterial $N_2$ fixation (Correll 1998; Lewis et al. 2011; Schindler et al. 2008). However, the results of nutrient enrichment experiments at a range of scales, from bottle bioassay to whole lake, have shown that the mass growth of phytoplankton and the formation of HABs are controlled by enrichment of both N and P, rather than N or P alone (Chen et al. 2013; Lewis and Wurtsbaugh 2008; Lewis et al. 2011; Paerl et al. 2016; Xu et al. 2015). The limiting roles of N and P have been found to vary temporally across seasons, geographically with regions, and even spatially within a lake. P limitation is commonly observed in spring, whereas it often changes to N limitation in summer and fall, when temperature and meteorological conditions favor the formation of HABs (Bullerjahn et al. 2016; Chaffin et al. 2013; Janssen et al. 2017). The change in paradigm from P limitation to dual limitation is because cyanobacterial $N_2$ fixation cannot always fulfil the N requirement by lake ecosystems. Inputs of P can be sustained by the release of P from sediments, while N deficits often occur due to denitrification (Lewis et al. 2011; Paerl et al. 2016).

The release of P due to internal sediment loading has been well recognized (Janssen et al. 2017; Lepori and Roberts 2017; Paytan et al. 2017; Xie et al. 2003), with P release persisting for typically 5–15 years after the reduction of external P inputs (Jeppesen et al. 2005; Watson et al. 2016; Welch and Cooke 2005). The rate of sediment P release can vary seasonally, with increases normally observed in warm seasons (Spears et al. 2012; Yang et al. 2013). It has been reported that in some lakes the contribution of sediment P release into the water column exceeds the external P input and becomes a critical factor influencing HABs (Nürnberg and LaZerte 2016; Penn et al. 2000; Sondergaard et al. 2003; Wu et al. 2017). In some cases, internal diffusive recycling of P cannot trigger HABs on its own, but can cause blooms when combined with increased external P loads (Matisoff et al. 2016). However, there are few experimental evidences to prove the theory that internal P loading from sediments can induce imbalance in N and P concentrations, and that the seasonal variation changes from P limitation to N limitation (Orihel et al. 2015).

In this study, a semi-enclosed bay in Lake Taihu was selected to study the effect of internal P loading on seasonal nutrient limitation in the water column. High-resolution dialysis (HR-Peeper) and diffusive gradients in thin films (DGT) samplers were used to measure the distributions of mobile P and Fe(II) in sediments. The diffusion flux across the sediment-water interface (SWI) was calculated along with its contribution to the water column P, allowing the role of internal P loading on the formation of seasonal N limitation to be assessed.

## 2. Materials and methods

### 2.1. Lake description and field site

Lake Taihu is located in the southeastern region of the Yangtze River delta, in China's coastal plain (Fig. 1). It is a large and shallow lake, with a mean depth of around 2.0 m, covering a 2340 km$^2$ area. Harmful algal blooms were first reported in a few regions of Lake Taihu in the 1960s (Qin et al. 2004). The impacted regions gradually expanded to most of the lake by the mid- to late 1990s (Xu et al. 2017). During HAB events, cyanobacteria have been found to account for the majority of phytoplankton biomass (60–90%), based on Chl*a* concentrations (Otten et al. 2012; Xu et al. 2017). In May 2007, a highly publicized drinking water crisis occurred, in which the Wuxi city drinking water plant ceased functioning due to a very large "cyanobacteria mat" (Qin et al. 2010). Following this, a wide range of measures were implemented by Chinese central and local government to reduce external nutrient loading to the lake (Wu and Hu, 2008; Yang and Liu 2010). As a result, the concentrations of TN and TP in Meiliang Bay consistently decreased after 2007, returning to the level observed in the early of 1990s (Xu et al. 2017). However, Chl*a* concentrations did not respond with the reductions of TN and TP as anticipated.

The sampling site is located in Meiliang Bay nearby the Taihu Laboratory for Lake Ecosystem Research (TLLER) (31°26′18″ N, 120°11′12″ E), Nanjing Institute of Geography and Limnology (Fig. 1). Meiliang Bay is one of the most eutrophic regions of Lake Taihu (Xu et al. 2014). The annual Chl*a* concentration in Meiliang Bay continued to increase to a peak of 43 μg/L from 2007 to 2009, remaining over 20 μg/L following this (Xu et al. 2017). The three rivers connecting to Meiliang Bay, Wujing Gang, Zhihu Gang and Liangxi River, have all been closed off by local government in order to prevent new input of external sewage into the bay.

### 2.2. Principles and preparation of HR-peeper and DGT samplers

The HR-Peeper device contained 30 equally spaced 200 μL chambers fully loaded with deionized water, with a 4.0 mm vertical resolution (Ding et al. 2010; Xu et al. 2012) and the chamber surfaces were covered using a 0.45 μm cellulose nitrate membrane. The concentration of soluble analytes present in sediment pore water, was measured by analyzing sample solutions in chambers after equilibration.

DGT is a passive sampling technique for the measurement of the labile fraction of analytes (Zhang et al. 2014; Zhang and Davison 1995). Concurrently, high-resolution DGT measurements were performed at

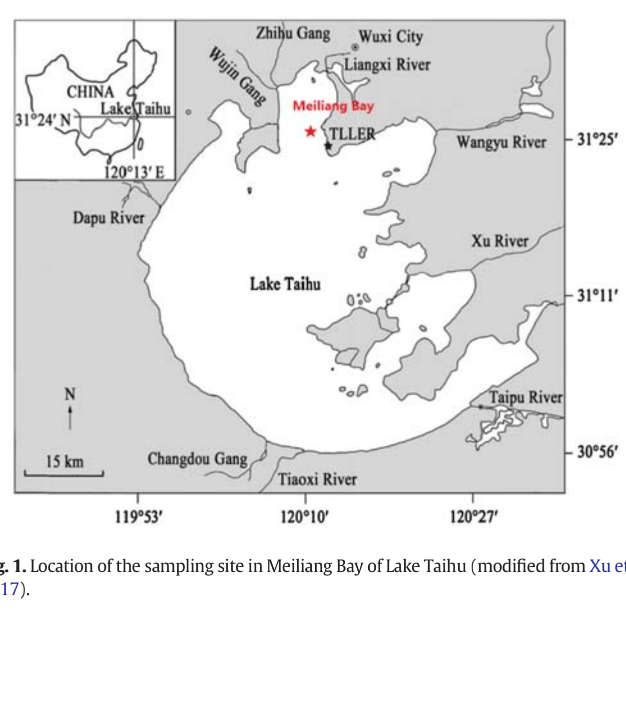


**Fig. 1.** Location of the sampling site in Meiliang Bay of Lake Taihu (modified from Xu et al. 2017).

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